BEHAVIOUR OF THERMAL EXPANSION OF THE $(1-x)Pb(Ni_{1/3}Nb_{2/3})O_3 - xPbTiO_3$ SOLID SOLUTIONS

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Abstract

The features of the dependence of the bulk coefficient of thermal expansion of the (1-x) $Pb(Ni_{1/3}Nb_{2/3})O_3$-$xPbTiO_3$ solid solution studied in series of x = 0 – 0.8 in the range from 100 to 750 K are reported. The obtained data and observed anomalies of thermal expansion are compared with dielectric properties, heat capacity and the phase diagram of concentration. The anomalous and lattice constituents of deformation and of the coefficient of thermal expansion are analysed the data on deformation being used to estimate the Burns' temperature and the mean-square polarisation of the samples found to decrease with the decrease of $PbTiO_3$ concentration.

Key words: Ferroelectrics, relaxor, ceramics, solid solutions, thermal expansion, phase diagram.

1. Introduction

Growing attention lately is paid to multicomponent mixed oxide perovskites of odd crystalline structure and unique physical properties – an enormous dielectric permittivity, large piezoelectric and electrostriction coefficients in particular. The (1-*x*)PNN–*x*PT solid solutions of the $Pb(Ni_{1/3}Nb_{2/3})O_3$ (PNN) relaxor and ferroelectric $PbTiO_3$ (PT) are a remarkable example [1-4].

PNN has its place in the family of oxide relaxors such as the synthesised at the same time well-known $Pb(Mg_{1/3}Nb_{2/3})O_3$ (PMN) [5] and is a multiferroic. A broad frequency-dependent maximum of the dielectric permittivity ε in PNN is observed at $T_m \approx 153$ K, which is much lower than in the PMN and likely the reason of PNN being less studied. The antiferromagnetic transition in PNN proceeds at $T_N \approx 5$ K [6].

Ferroelectric lead titanate being known since long ago is still actively studied [7, 8]. Transition between the paraelectric and ferroelectric phases proceeding in $PbTiO_3$ at $T_c = 763$K is characterised as a first-order structural transformation with the symmetry change $Pm3m \rightarrow P4mm$ occurring close to the tri-critical point.

Structural and electrical studies of the (1-*x*)PNN–*x*PT system [1, 9-12] have shown the phase transition temperature characteristic to PT decreasing with the decrease of *x*. A morphotropic region exists on the *T*–*x* phase diagram around $x \approx 0.35$ wherein several phases can come about – the cubic, pseudo-cubic, tetragonal and rhombohedral [9].

More recent studies of heat capacity [13-15] have revealed additional anomaly of uncertain nature at ~520 K apart from the anomalies characteristic to phase transitions (at $x > 0.35$) and relaxor behaviour at T_m ($x < 0.35$). Presently the crystal structure and its change with the temperature is a subject of discussion. All of that stimulates further detailed studies of the mixed ferroelectric relaxors.

The paper presents results of a study of thermal expansion of the (1-*x*)PNN–*x*PT ($x = 0$–0.8) ceramics in the range from 100 to 750 K. Thermal expansion being related to properties (ferroelectricity, piezoelectricity and pyroelectricity) is one of the basic characteristics of the material. Thermal expansion is important as well for design of technological devices as for

uncovering the physical nature of phenomena observed in solid solutions, the crossover from the usual ferroelectric to relaxor behaviour. Polarisation and its transformation with the change of composition have also been determined from the data of thermal expansion since often the use of traditional techniques in the case of ceramic materials is complicated. Deformation mainly being determined by polarisation square can be used for assessment of the macroscopic spontaneous polarisation of ferroelectrics and the mean square of polarisation in relaxor ferroelectrics.

2. Samples and methods

2.1. Samples

The samples were prepared in the Institute of Solid State Physics of the University of Latvia by conventional ceramics technology [13, 14] from oxides: 99.5 % purity grade PbO, 99.0 % purity grade NiO, 99.9 % – Nb_2O_5, and 99.9 % – TiO_2.

The primary mixture in accordance to composition and mass of the matching oxides was processed for 12 hours with alcohol in jasper mills, then dried for 6 hours before being pressed into ingot tablets subject to solid phase calcinating at 900 °C for 1-3 hours the technological parameters depending on the batch of the initial ingredients being optimised separately for each of the compounds. After subsequent crushing of the tablets and homogenising the calcinated mass by milling it was pressed again into tablets to fire ceramics at 1150–1270 °C for one to three hours. After a slow cooling of the synthesized tablets and removal of the surface layer the samples were cut and polished for measurements.

Conditions of the thermal treatment were optimised to improve the structure and to surge the perovskite phase in the material. The quality and the phase composition was studied by X-ray analysis of the synthesised material and the fired ceramics revealing the stoichiometric perfectness of the obtained compounds as well as presence of the undesired pyrochlore phase and other admixtures. The technological parameters used in the study are presented in Table 1.

2.2. Thermal expansion

Thermal expansion of ceramic samples of $L \approx 5–10$ mm was studied in dynamic process by use of an NETZSCH DIL-402C induction dilatometer within the 100–750 K range at the rate of heating and cooling of 2–3 K/min. The measurements were made in the flow (~50 ml/min) of dry helium (concentration of O_2 not exceeding 0.05 % in bulk) at the load of 30 cN applied to the sample the calibration of the measuring system being made by melted quartz standards.

Several series of measurements were conducted with different samples of the same composition. Since, as a rule, the data between the series agreed within the accuracy of the measurements, they were processed together.

3. Results and discussion

3.1. The data of measured thermal expansion

Dependences of the coefficient of the bulk thermal expansion $\beta = 3\alpha$ and the bulk deformation $\Delta V/V$ of the (1-x)PNN–xPT ceramics are presented in Fig. 1 and Fig. 2 together with the results of thermal expansion of PT [16] from earlier studies.

The anomaly of the bulk coefficient of thermal expansion $\beta(T)$ at the cubic-to-tetragonal phase transition (at T_c) characteristic to pure PT is found to remain distinct enough while of remarkably decreasing amplitude in the (1-x)PNN–xPT solid solutions with the decrease of x down to 0.5. At concentrations of PT below the morphotropic phase boundary at $x \approx 0.35{-}0.38$ the anomaly broadens reaching its minimum at T_m and its shape becomes characteristic to ferroelectric relaxors (Fig. 1).

Table 1. Technological parameters at baking the studied samples of (1-x)PNN–xPT ceramics.

Composition	*Sample*	*Calcin. $^0C/h$*	*Synth. $^0C/h$*	*Density g/cm^3*	*Cell Å*	*Addition*
PNN	0.0A		1150/2	7.6176	cubic. a=4.03009(7)	
0.9PNN-0.1PT	0.1A	900/3	1170/3	7.9422		9% pyrochlore
0.9PNN-0.1PT	0.1B	900/3	1210/1	7.3286	cubic, a=4.02434(24)	7% pyrochlore 10% NiO
0.8PNN-0.2PT	0.2A	900/3	1230/1			
0.8PNN-0.2PT	0.2B	900/3	1200/4		tetragonal, close to cubic a=4.01430(28) c=4.02664(46)	5% NiO
0.7PNN-0.3PT	0.3A	900/3	1200/1			
0.7PNN-0.3PT	0.3B	900/3	1200/1			
0.65PNN-0.35PT	0.35A	900/3	1230/1	8.1153	tetragonal, a=3.99796(10) c=4.01755(13)	1% 3.6% NiO
0.65PNN-0.35PT	0.35B	900/3	1260/1		tetragonal	
0.6PNN-0.4PT	0.4A	900/3	1230/1	8.0734	tetragonal a=3.950202(98) c=4.05699(15)	pyrochlore 1% NiO 1.3%
0.6PNN-0.4PT	0.4B	900/3	1260/1		tetragonal	
0.5PNN-0.5PT	0.5A	900/3	1270/1		tetragonal a=3,97674(78) c=4,03261(91)	2% pyrochlore
0.5PNN-0.5PT	0.5B	900/3	1270/1	7.6021	tetragonal a=3,97674(78) c=4,03261(91)	2% pyrochlore
0.4PNN-0.6PT	0.6A	900/3	1250/1	7,8739	tetragonal a=3,96280(49) c=4,04513(50)	1-2% addition
0.3PNN-0.7PT	0.7A	900/3	1230/1	7.6739	tetragonal a=3,949457(77) c=4,05875(12)	1% pyrochlore
0.3PNN-0.7PT	0.7B	900/3	1260/1			
0.2PNN-0.8PT	0.8A		1230/1	7,6580	tetragonal. a=3,93218(40) c=4,07805(46)	
0.2PNN-0.8PT	0.8B	900/3	1250/1	7,4555	tetragonal a=3,93336(29) c=4,07845(37)	

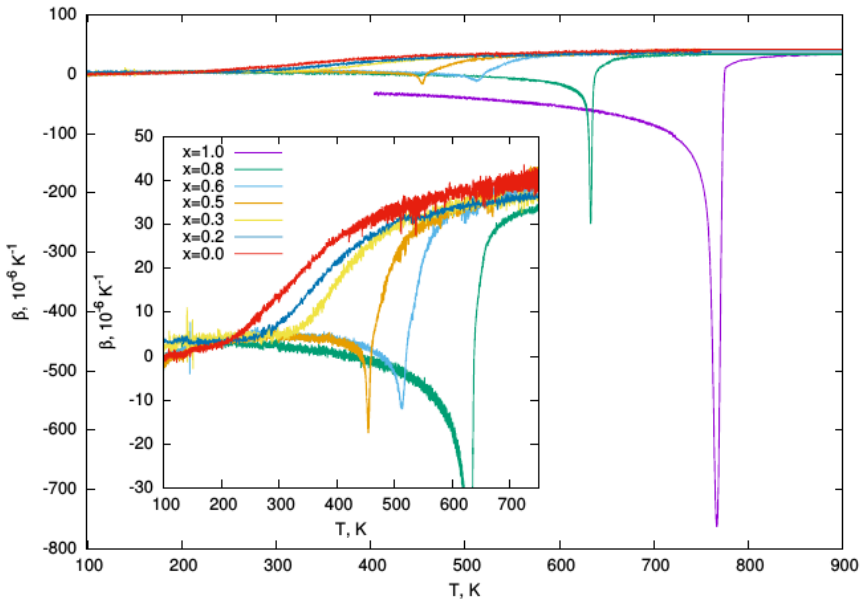

Fig.1. Dependence of the bulk coefficient of thermal expansion of the samples of (1-x)PNN–xPT ceramics on temperature. The data on pure PT from [16].

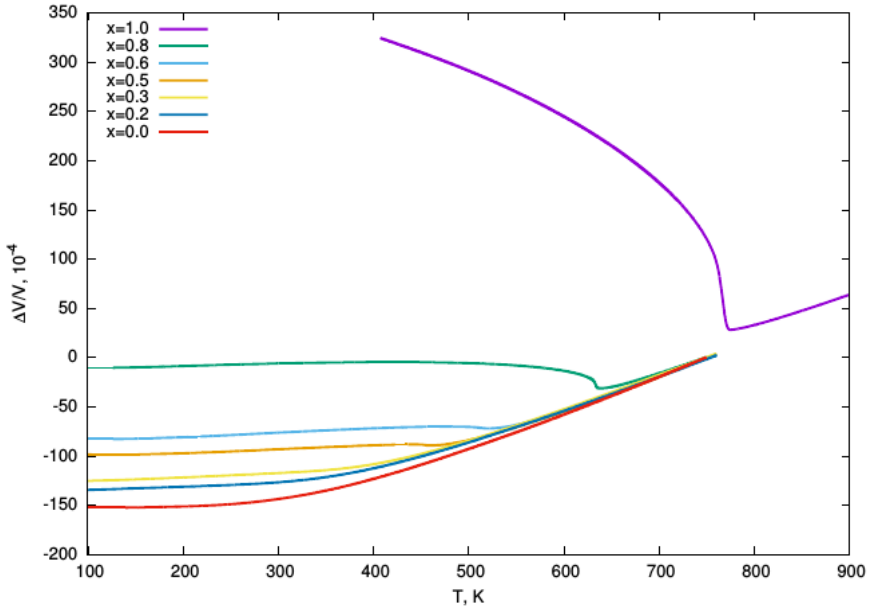

Fig. 2. Dependence of bulk deformation ΔV/V of the samples of (1-x)PNN–xPT ceramics on temperature. The data on pure PT from [16].

3.1.1. Additional anomalies of thermal expansion

In earlier studies [13-15] of heat capacity of the (1-*x*)PNN–*x*PT ceramics of *x* = 0.5, 0.4, 0.3 anomalies characteristic to phase transitions of undefined nature have been observed at ~520 K. We also have detected small additional anomalies of thermal expansion in some samples of *x* = 0.2, 0.3, 0.6 and 0.8.

Anomalous behaviour of $\beta(T)$ was observed in some samples of x = 0.8 (0.8B) at $T \approx 645$ K above T_c (Fig. 3) while the basic anomaly had a smaller amplitude and was shifted to a lower temperature as compared with the sample of the same composition (0.8A) without additional anomalies.

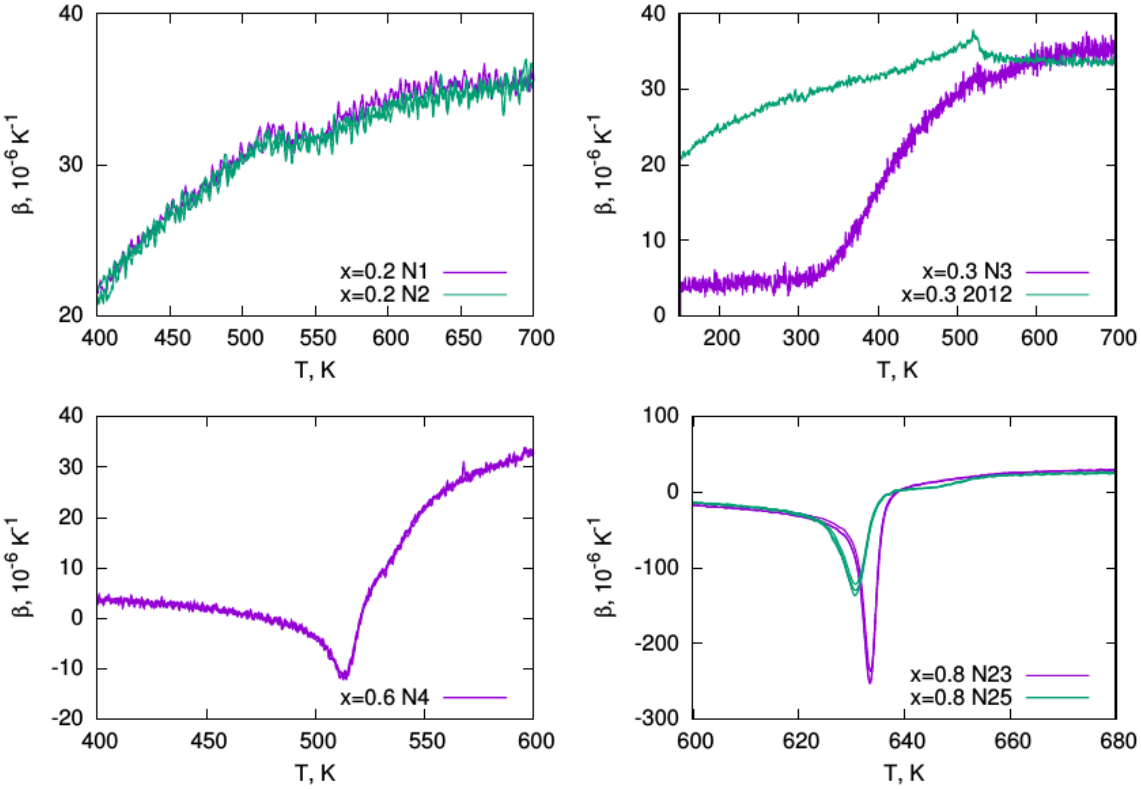

Fig. 3. Dependence of the bulk coefficient of thermal expansion of the samples of (1-x)PNN–xPT ceramics of x=0.2; x=0.3; x=0.6; x=0.8 on temperature.

The same is likely observed in sample 0.6A of x = 0.6 in which the anomaly is somewhat reduced with regard to what would be expected from the trend of β at decreasing x (Fig. 1). In compounds of x = 0.2 and 0.3 an anomaly observed in heat capacity [13] is seen at $T \approx 520$ K. Dependencies of the coefficient of thermal expansion in two samples of x=0.3 (0.3A and 0.3B) are strongly different. While in sample 0.3B behaviour of β(T) is consistent with other compositions (x=0 and 0.2) the broad minimum of β(T) characteristic to relaxors practically is not observed in sample 0.3 A exhibiting the most pronounced additional anomaly.

The reason of such behaviour is likely related to conditions under which the samples are prepared, to thermal history and to possible presence of foreign phases. The main problem with studies and application of the PNN and PNN-PT ceramics [17], as well as PMN and PMN-PT ceramics [18] is the trouble of obtaining a single-phase perovskite material. The problem of the synthesis is the undesired formation of the pyrochlore phases – $Pb_{1.45}Nb_2O_{6.26}$ or $Ti(Ni_{1/3}Nb_{2/3})O_4$ [19].

Results obtained on samples without or smallest additional anomalies Figs. 1 and 2) are used for farther analysis.

3.2. The phase diagram of concentration

The T–x phase diagram of the (1-*x*)PNN-*x*PT system is presented in Fig. 4 together with data of the dielectric [9] and thermal capacity [13 - 15] studies. Within the studied range of concentration and the measurement accuracy the values of T_c and T_m are in good agreement with data published by other authors.

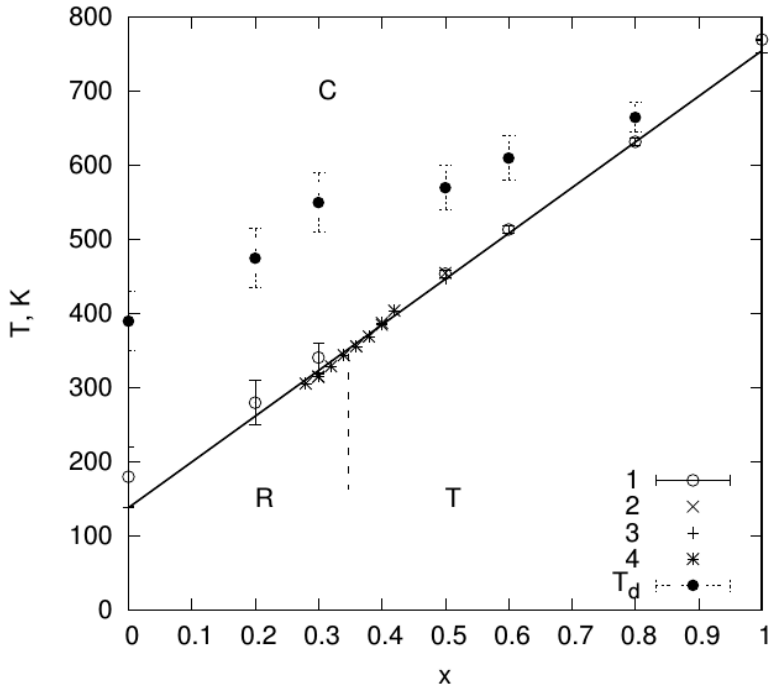

Fig. 4. The phase diagram of concentration of the (1-x)PNN-xPT system

3.3. Anomalous contributions to thermal expansion

3.3.1. Thermodynamics

In $PbTiO_3$ and its solid solutions, as in other perovskites, the ordering parameter is presented by spontaneous polarisation **P** = (P_1,P_2,P_3). Deformation determined by the square of the macroscopic spontaneous polarisation can be used to estimate the latter:

$$\begin{aligned} e_{11} &= Q_{11}P_1^2 + Q_{12}P_2^2 + Q_{12}P_3^2, \\ e_{22} &= Q_{12}P_1^2 + Q_{11}P_2^2 + Q_{12}P_3^2, \\ e_{33} &= Q_{12}P_1^2 + Q_{12}P_2^2 + Q_{11}P_3^2, \end{aligned} \qquad (1)$$

$$\Delta V/V = e_{11} + e_{22} + e_{33} = (P_1^2 + P_2^2 + P_3^2)(Q_{11} + 2Q_{12}) = P^2(Q_{11} + 2Q_{12}) \qquad (2)$$

where Q_{11} and Q_{12} are the coefficients of electrostriction.

It is necessary to have a correct description of the non-anomalous contributions to deformation and the coefficient of thermal expansion $\beta_L(T)$ to separate the anomalous contribution of deformation and to determine the polarisation P.

3.3.2. Separation of the anomalous contributions

To separate the anomalous contribution to deformation and to assess the mean square of polarisation in relaxors the deformation is assumed, as a rule, being linear at high temperatures ($T > T_c$, $T >> T_m$) [20 - 23].

Since the $\Delta V/V(T)$ is obviously nonlinear and the coefficient of thermal expansion $\beta(T)$ is not constant, such approach does not provide an adequate description of experimental data away from the phase transition to low temperatures. Amounts of the anomalous contributions to deformation and mean square polarisation determined this way are, as a rule, overestimated and depend on the range of temperature where the approximation is made.

The coefficient of thermal expansion β slightly though depends on the temperature above the Debye temperature which is the reason of nonlinearity of the deformation. Besides, due to thermo-stimulated defects there are additional contributions to deformation at high temperatures as evidenced by the behaviour of the coefficient of thermal expansion going to zero with decreasing the temperature. At low temperatures it is necessary to account for the interconnection between thermal expansion and heat capacity along with its dependence on the temperature at least within the Debye (or the combined Debye-Einstein) model.

The data on the dependence of the regular (lattice) component of the coefficient of the bulk thermal expansion β(T) were processed by use of

$$\beta_L(T) = aT + bC_D(\Theta_D, T) + cC_E(\Theta_E, T), \qquad (3)$$

where

$$C_D(\Theta_D, T) = 9R\left(\frac{\Theta_D}{T}\right)^3 \int_0^{\Theta_D/T} t^4 \frac{\exp(t)}{(\exp(t)-1)^2} dt, \qquad (4)$$

$$C_E(\Theta_E, T) = 3R\left(\frac{\Theta_E}{T}\right)^2 \frac{\exp(\Theta_E/T)}{(\exp(\Theta_E/T)-1)^2}, \qquad (5)$$

a, *b*, *c*, Θ_D, Θ_E – fitting parameters.

3.3.3. The anomalous contribution to deformation and estimation of the Burns temperature

Dependencies of the anomalous contribution to the bulk deformation on the temperature calculated from

$$\Delta V/V - \int \beta_L(T) dT = (Q_{11} + 2Q_{12})P^2, \qquad (6)$$

are presented in Fig. 5. A nonzero contribution is observed in all samples above the phase transition temperature T_c or T_m, which is related to emerging polar nano-regions of random orientation below the Burns' temperature T_d.

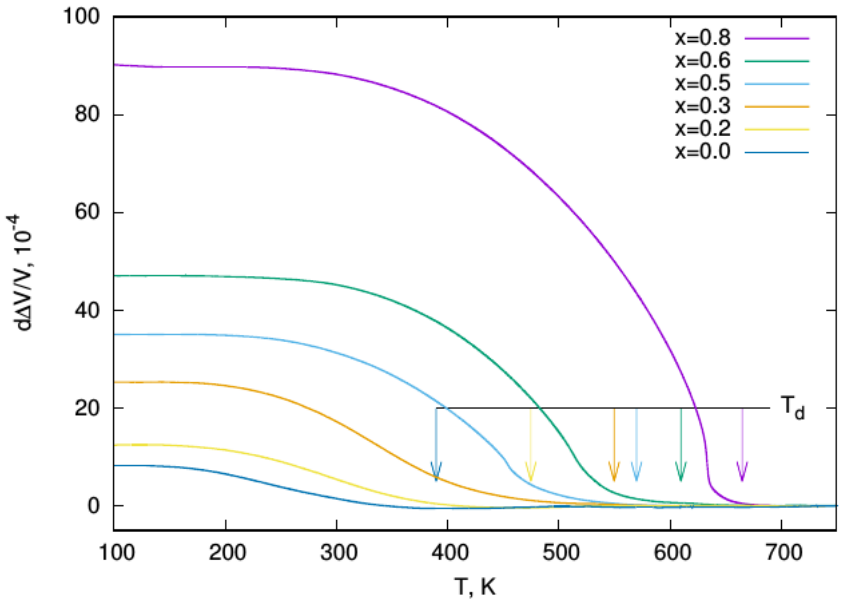

Fig. 5. The anomalous contribution to bulk deformation as a function of temperatuure.

The macroscopic spontaneous polarisation P_r above the temperature of transition to the ferroelectric state is equal to zero while the mean square polarisation in the polar nano-regions system is not zero ($P_d = <P^2>^{1/2} \neq 0$). The latter dynamic polarisation responsible for a contribution into the anomalous thermal expansion can be found from it. The calculations of P_d were made employing the electrostriction coefficients of pure PT (Q_{11} =8.9×10^{-2} m^4/C^2, Q_{12} =-2.6×10^{-2} m^4/C^2) assuming they do not change much by adding PNN. Dependence of P_d on temperature in the studied samples is shown in Fig. 6. The values of saturated P_d (at 100 K) and other characteristics of the ceramics obtained from thermal expansion data along with the spontaneous macroscopic polarisation P_r from P-E loops of the compounds of x= 0.4 [3, 4] are presented in Table 2.

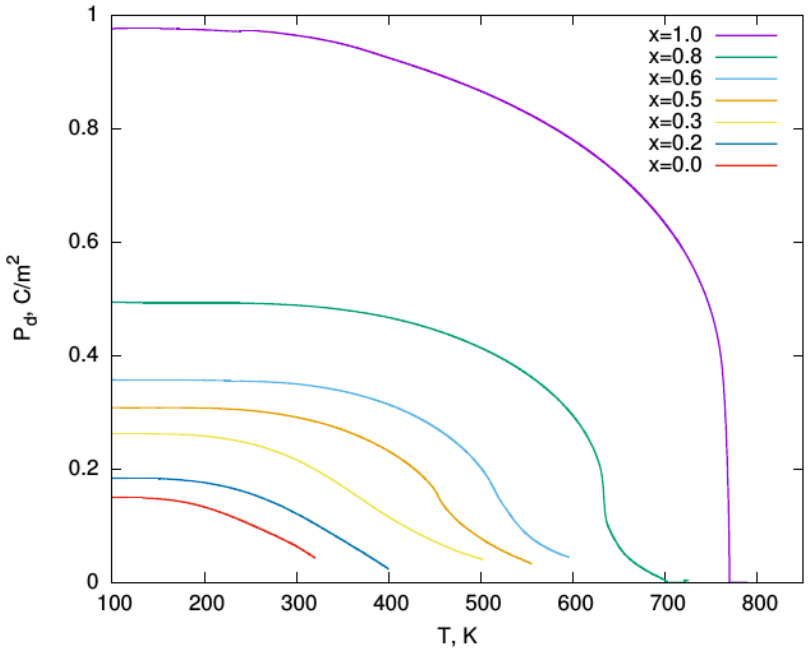

Fig. 6. The mean square polarisation P_d of the studied (1-x)PNN–xPT compounds.

Table 2. Characteristics of the (1-x)PNN–xPT ceramics system obtained from the data of thermal expansion.

Compound	T_c, **K**	T_m, **K**	T_d, **K**	P_d, **μC/cm^2**	P_r, **μC/cm^2**
PNN		180±30	390±40	15	
0.8PNN-0.2PT		280±30	475±40	18	
0.7PNN-0.3PT		340±20	550±40	25	
0.6PNN-0.4PT	400				30 [4]; 18 [3]
0.5PNN-0.5PT	454±5		570±30	32	
0.4PNN-0.6PT	514±5		610±30	36	
0.2PNN-0.8PT	632±5		665±20	47	
PT	770±1			96; 86 [19]	40-90 [21,22]

Pure lead titanate has been studied long since and scrupulously. Abundance of diverse information on physical and structural properties allegedly allows regarding it as a model ferroelectric material. However, a number of reasons make it problematic to agree on the details of the phase transition mechanism. For instance, obtaining of well-sintered polycrystalline samples presents a severe problem because of stoichiometry being defied by volatility of lead oxide at high temperatures [7]. Crystals grown by various techniques even at room temperature have considerable conductivity intervening with studies of dielectric properties [24, 25] and evidently cause substantial ambiguity in values of saturated polarisation over the range of P_r =(40–90) μC·cm^{-2} measured by different authors [24, 25]. The value of polarisation of PT estimated from the data on thermal expansion in the present study is in good agreement with the data published by other authors [22, 24, 25].

The data available on polarisation in the (1-x)PNN–xPT solid solutions [3, 4] and in pure PT are of the order of magnitude consistent with the present results confirming reliability of the employed method.

4. Conclusions

The anomaly of the bulk coefficient of thermal expansion at the transition from the cubic to the tetragonal phase is found to stay distinct enough while its intensity is decreasing with the decrease of *x*. At PT concentrations below the morphotropic boundary the anomaly broadens acquiring the pattern specific to relaxor ferroelectrics.

The Burns' temperature T_d at which the polar nano-regions emerge is determined along with T_c and T_m from the data of thermal expansion of the *(1-x)PNN–xPT* solid solution series studied in samples of x = 0, 0.2, 0.3, 0.5, 0.6 and 0.8.

The interval between T_d and T_m reaching ~200 K in compounds of $x < x_{MPB}$ decreases with the growth of PT concentration. The possibility to separate the regular and anomalous contributions to deformation and reliably estimate the mean polarisation square P_d is confirmed by analysis of the obtained results.

The local mean square polarisation present in *(1-x)PNN–xPT* compounds decreases with the decrease of PT concentration.

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Legends

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Fig. 2. Dependence of bulk deformation $\Delta V/V$ of the samples of (1-x)PNN–xPT ceramics on temperature. The data on pure PT from [16].

Fig. 3. Dependence of the bulk coefficient of thermal expansion of the samples of (1-x)PNN–xPT ceramics of x=0.2; x=0.3; x=0.6; x=0.8 on temperature.

Fig. 4. The phase diagram of concentration of the (1-x)PNN-xPT system.

Fig. 5. The anomalous contribution to bulk deformation as a function of temperatuure.

Fig. 6. The mean square polarisation P_d of the studied (1-x)PNN–xPT compounds.